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14. ABSTRACT
A new measurement technique has been proposed to determine the sign of the index of refraction in thin film samples.
We have observed signature of negative index in NiFe-SiO2 magnetic granular materials and in NiFe/SiO2 multilayers. However, the signal is weak due to thin sample and is very much sample dependent, we could not consistently confirm the properties.
We have theoretically established selection criteria for magnetic materials and their structures to achieve LHMs or NIMs.
We have theoretically proposed several new structures that show negative index of refraction (NIMs). These structures include:
1. Double negative materials (DNMs) for LHMs: E/M multilayers consisting of alternating negative e and negative u layers.
2. Single negative materials (SNMs) for NIMs: Ferrite/(Semiconductor or Oxides) multilayer with negative u.
We have developed a theory that unifies DNMs and SNMs as a function of two flmdamental material parameters: quality factors for permittivity (Qe=e'/e") and permeability (Qu=u'u").

15. SUBJECT TERMS

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June 1, 2003 to August 31, 2006

Left Handed Materials Based on Magnetic Nanocomposites

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Objective

The main objectives of this project are:

- Develop LHM based on magnetic composites
- Demonstrate special EM properties that have potential DoD applications

The proposal was based on the postulation that negative ε automatically occurs in metallic magnetic entities for frequencies less than the plasma frequency, which typically occurs in the UV range, and a negative magnetic permeability μ can be achieved at the vicinity of ferromagnetic resonance. By choosing different material and structure combinations, we may obtain simultaneously negative effective permeability $\mu_{\rm eff}$ and permittivity ε_{eff} to form LHM or only single negative parameter (SNM) to form negative indexed materials (NIM).

Achievement:

- A new measurement technique has been proposed to determine the sign of the index of refraction in thin film samples.
- We have observed signature of negative index in NiFe-SiO_2 magnetic granular materials and in NiFe/SiO_2 multilayers. However, the signal is weak due to thin sample and is very much sample dependent, we could not consistently confirm the properties.
- We have theoretically established selection criteria for magnetic materials and their structures to achieve LHMs or NIMs.
- We have theoretically proposed several new structures that show negative index of refraction (NIMs). These structures include:
    1. Double negative materials (DNMs) for LHMs: E/M multilayers consisting of alternating negative ε and negative μ layers.
    2. Single negative materials (SNMs) for NIMs: Ferrite/(Semiconductor or Oxides) multilayer with negative μ.
- We have developed a theory that unifies DNMs and SNMs as a function of two fundamental material parameters: quality factors for permittivity ($Q_\varepsilon = \varepsilon'/\varepsilon''$) and permeability ($Q_\mu = \mu'/\mu''$).

In the following we will detail the findings.

1. Experimental results in magnetic granular and multilayer films

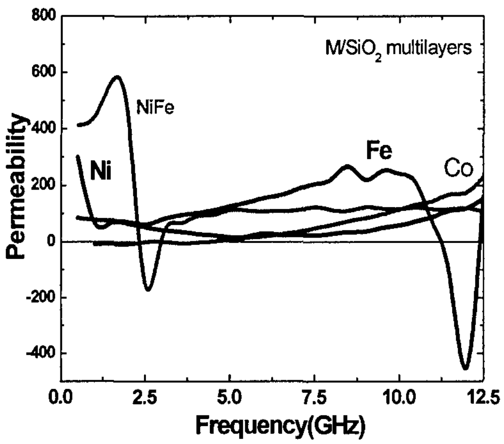

Figure 1, Ferromagnetic resonance of various magnetic materials in zero magnetic field.

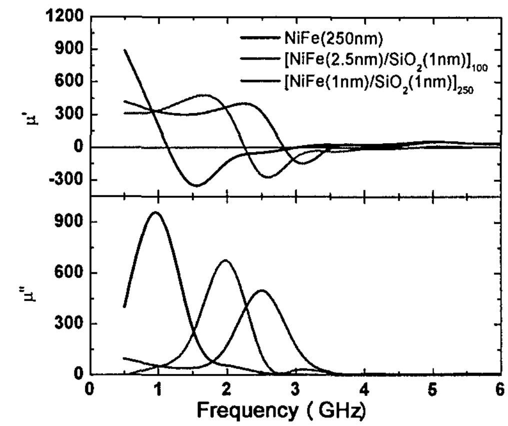

Figure 2, magnetic permeability μ'+jμ" of NiFe films of different thickness as a function of frequency. The total NiFe layer thickness is kept at the same at 250 nm.

As a first step, we have measured the ferromagnetic resonance ω_o in zero magnetic fields for common magnetic materials including FeNi, Ni, Co, and Fe (*Figure*). The value of ω_o is mainly proportional to the magnetocrystalline anisotropy k_1 and thus progressively increase from Ni (~ 1 GHz), FeNi (~ 2.3 GHz), Fe (~ 11.2 GHz), and Co (>12.5 GHz). In additional to k_1, the shape anisotropy and magnetic field can also be used to tune ω_o. The effect of shape anisotropy which can be changed by the film thickness as illustrated in *Figure* using NiFe/SiO2 multilayers. We have also measured permeability spectrum in granular (NiFe)-Si and NiFe-PSU (polystyrene-urethane) and observed negative permeability (*Figure 3*). From these measurements, it is clearly that FeNi, Fe, and Co are good magnetic materials for LHM at low, mediate, and high frequencies.

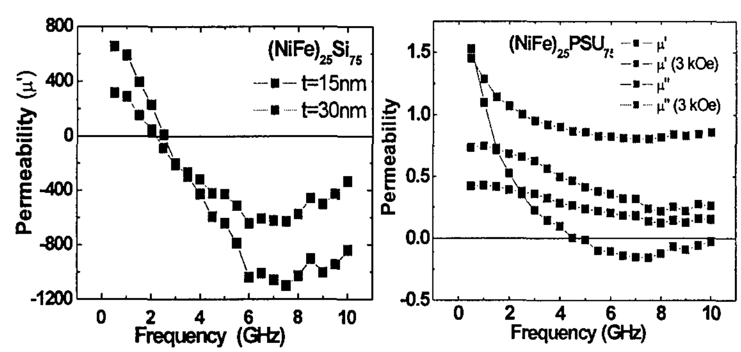

Figure 3, Magnetic permeability measured with a resonant cavity for 15 nm and 30 nm thick $(NiFe)_{25}Si_{75}$ (vol.%) thin films made by magnetron sputtering (left panel) and 4 mm thick $(NiFe)_{25}PSU_{75}$ (polystylenreurethane) sample made by the extrusion technique. In the latter, an external magnetic field is applied to achieve negative

Selection Criteria of Magnetic Materials

It is interested to understand the condition for negative permeability. For which we analyzed theoretically the bandwidth of negative permeability. Because the negative ε_{eff} is extended to the plasma frequency of a metal, which is typically in the UV region, the BW of m-LHM is predominately determined by the frequency width where negative μ exists. For magnetic materials, the BW for negative μ depends on the magnetic damping coefficient α and the characteristic frequencies ω_o and ω_m, where $\omega_m = \gamma M_s$. *Figure 4* shows the permeability dependence of damping constant. With zero damping, negative μ can be observed between ω_o and $\omega_o+\omega_m$, leading to ideal $BW = \omega_m$. However, the negative μ will disappear once it reaches the critical damping.

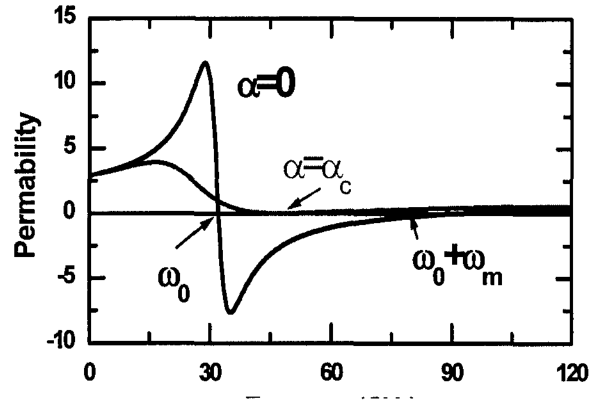

Figure 4, there is a critical damping above which negative permeability disappears

For soft magnetic materials with high magnetization such as NiFe, the ratio of ω_m/ω_o is about 2, leading to α_c around 0.5. Consequently, substantial BW can be observed as it can be seen in *Figure 5*. Ni has a small magnetization, leading to small ω_m/ω_o and α_c around 0.2. Therefore, a little damping can destroy the BW of negative permeability. For hard magnetic materials such as FePt and CoPt, the ratio of ω_m/ω_o is about 0.1, which leaves much smaller tolerant space for damping constant. Based on the above analysis, we choose NiFe as our primary candidate to achieve LHM on magnetic composites and magnetic metallic/dielectric multilayers.

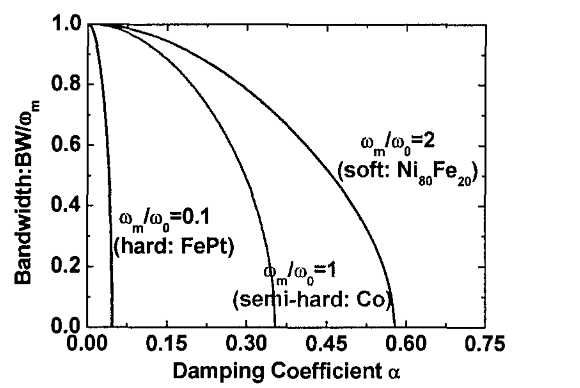

Figure 5, the negative permeability bandwidth (BW/ω_m) as a function of damping coefficient α for ω_m/ω_0 =2, 1, and 0.1, respectively

Novel experimental technique to measure index of refraction in thin film samples

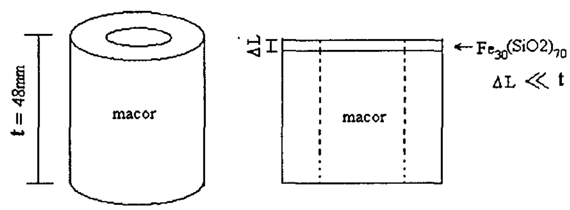

Figure 6, Left: Macor substrates. Right: $Fe_{30}(SiO_2)_{70}$ fabricated on top of Macor. The sample thickness ΔL is much smaller than the substrate thickness t.

Because of the nature of thin films (~ 100 nm), it is difficult to measure the index of refraction. We have developed a new experimental method to measure the sign of the index of refraction The method is to measure how magnetic samples shift the resonance peaks of the substrates in a coaxial line. A dielectric substrate of a finite thickness is shown in the left panel of **Error! Reference source not found.** It will create a series of resonant peaks (black curves in *Figure)*. These peak positions are shifted by the magnetic thin films deposited on top of the substrate (*Figure 6*- right panel) as shown in the red curves in *Figure* . The resonant peak shift is proportional to $n\Delta L$, where n and ΔL are the index of refraction and the thickness of the $Fe_{30}(SiO_2)_{70}$ film. If $n > 0$, these peaks will be shifted to lower frequencies, which is the case at $f < 12.5$ *GHz*. If $n < 0$, the peaks will be shifted to higher frequencies, as in the case of $f > 12.5$*GHz*. Therefore, the index of refraction n of $Fe_{30}(SiO_2)_{70}$ changes sign at about $f = 12.5$ *GHz*, suggesting it is a LHM at $f > 12.5$*GHz*. This transition frequency is very close to the resonant frequency of Fe films depicted in *Figure* (blue curves).

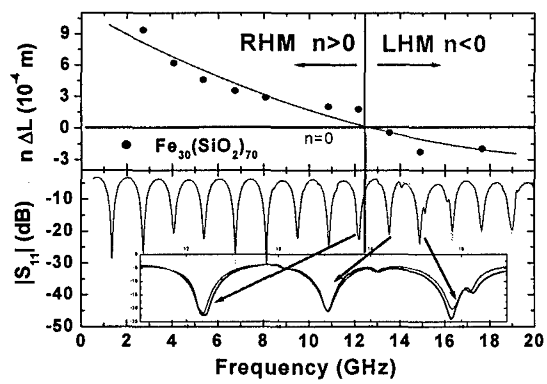

Figure 7, Reflectivity S_{11} of Macor (black curve) and $Fe_{30}(SiO_2)_{70}$/Macro (red curve). The top panel plots the product of n and $Fe_{30}(SiO_2)_{70}$ thickness ΔL, indicating the index of reflection n changes sign above 12.5 GHz, indicated by the vertical green line.

Figure 8 shows the result of granular $NiFe_{38}(SiO)_{26}$ nanocomposites. The resonance peaks shifted to higher frequencies between 2.4 GHz and 7.2 GHz, which indicates the sign of index of refraction is negative.

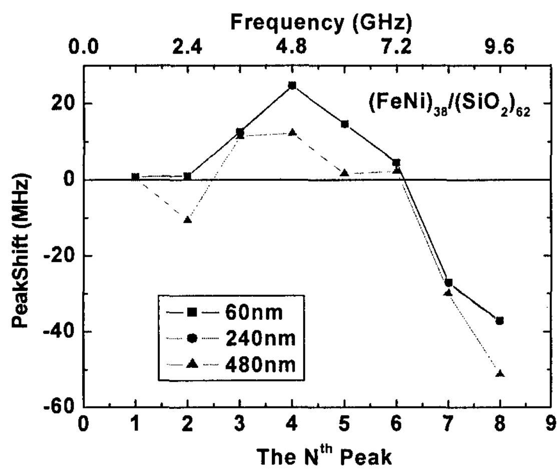

Figure 8, the resonance peak shift vs. frequency. It indicates the negative permeability between 2.4 GHz and 7.2 GHz

We can also determined the sign of index of refraction by looking the thickness dependent of the resonance peak shift. If the index of refraction is positive, the resonance peak continually shifts to lower frequency when the thickness increases. If the index of refraction is negative, the resonance peak continually shifts to higher frequency when the thickness increases. We can determine the sign of index of refraction by looking the peak shift vs. thickness relation. The positive slope of curve indicates the negative index of

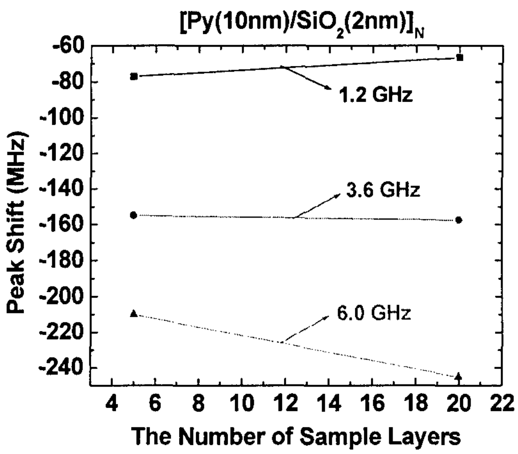

Figure 9, peak shift vs. thickness. The larger number of sample layers means the sample is thicker

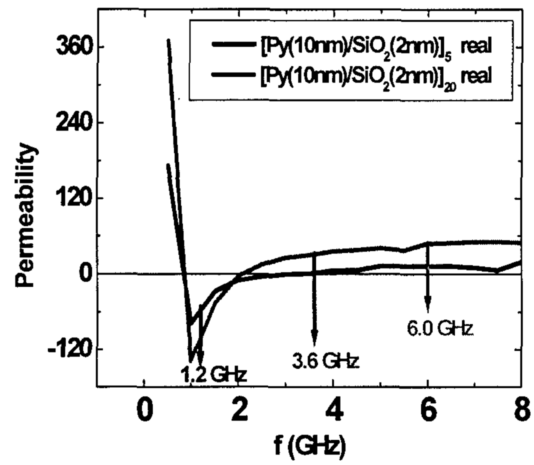

Figure 10, the microwave permeability of $[Py(10nm)/SiO_2(2nm)]_N$ multilayer

refraction, while the negative slope indicates the positive index of refraction. *Figure 9* shows the peak shift vs. thickness relation of $[Py(10nm)/SiO_2(2nm)]_N$ multilayers. A very clear transition between negative and positive index of refraction was observed. The index of refraction is negative at 1.2 GHz. Then it changes sign to positive at 3.6 GHz, and becomes a little larger positive at 6 GHz. We also measured the permeability of $[Py(10nm)/SiO_2(2nm)]_N$ multilayers, which is shown in *Figure 10*. The result is very consistent with the thickness dependent data where the negative index refraction is exactly in the negative permeability range.

While observed signals suggest the negative refraction in magnetic/insulator multilayers, the results are very sample dependent. We could not consistently reproduce our results. One main reason is, with our current facilities, we could not fabricate thick multilayer samples (> 10 μm) while precisely maintain the individual layer thickness. A proposal to request a pulse e-beam gun was not successful. Nevertheless, we strongly believe negative index of refraction exist in magnetic composites.

2. Proposed New Structure for LHMs and NIMs

During the program period, we have proposed several different combinations of material and structure to achieve LHM or NIM.

1. E/M multilayer consisting of alternating negative ε and negative μ layers

We can also achieve LHM using multilayers consisting of alternating negative ε and negative μ layers, dubbed as E/M multilayers. Negative ε can be achieved in any conducting layer and negative μ can be achieved in magnetic materials such as ferrites. By splitting the unit cell into two thickness layers (E and M layers) perpendicular to the direction of propagation, we have calculated the transmission and the reflection for normal incidence. The obtained effective $n_{eff}(\omega)$ and $\mu_{eff}(\omega)$ for the homogeneous effective medium approximation using the ISU retrieval procedure developed by Dr. Soukoulis are shown in *Figure 11*. The negative index refraction certainly can be obtained. However, it is difficult to fabricate the film since the thickness ratio between ferrite and metal is too large.

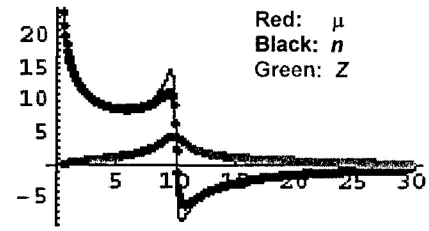

Figure 11, We consider the TE mode and a symmetric unit cell: (ε/2)(μ)(ε/2). The Ag layer is 1 angstrom, magnetic layer is 500 μm. The magnetic resonance has amplitude of 150

2. Ferrite/Semiconductor multilayer

We further investigated other possibilities of using semiconductors instead of metal layers, which have positive ε. Using the transfer matrix method, we determine that negative index refraction can still be achieved in ferrite/semiconductor multilayers. The result is shown is *Figure 12*.

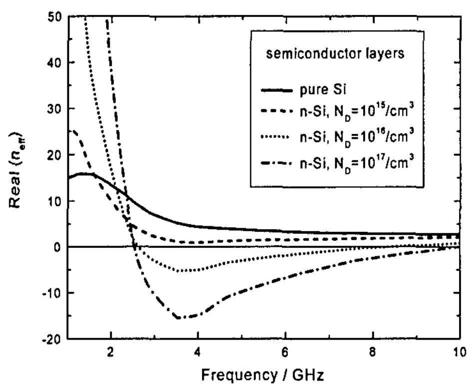

Figure 12, Real part of refraction index as a function of frequency for the layered silicon/CoBa ferrite film multilayers. The thicknesses are 200 nm and 1 μm respectively

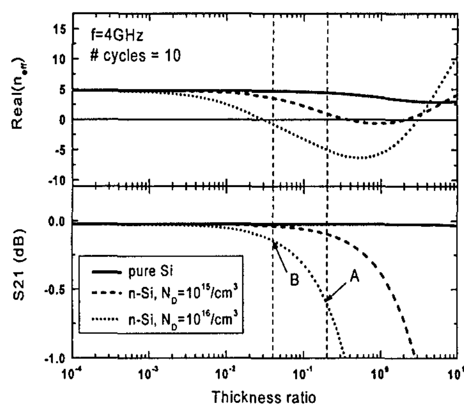

Figure 13, The real part of refraction index and the losses as a function of thickness of silicon films for silicon-BaCo ferrite film composite. Thickness ratio is defined as h_2/h_1. The loss is presented by scattering parameter S21.

The negative indices are found in certain impurity concentrations at a given film thicknesses. We also find that the magnitude of negative n_{eff} increases with increasing carrier density N_D. The frequency bandwidth for negative n_{eff} is confined in the frequency range where the BaCo ferrite has negative μ_r, and reduces with decreasing N_D. The negative μ_r of the ferrite provides the

composite an effective negative permeability, and therefore the frequency band is dominated by that of the ferrite materials. The negative n_{eff} depends not only on the impurity concentration but also on the thickness of silicon films. In *Figure 13*, we show the value of n_{eff} as a function of the silicon thickness (the thickness ratio h_2/h_1) for a given N_D at 4 GHz. The figure shows that the negative n_{eff} exists in certain range of thickness h_2 for doped silicon, and this range also relates to its impurity concentration. For example, the composite does not have any negative indices as shown in *Figure 13* when the thickness and the impurity concentration of the silicon films are 200nm (thickness ratio 0.2) and $10^{15}/cm^3$, respectively. But negative indices appear at 4 GHz when the thickness increases to 400 nm. For higher impurity concentrations, the negative indices will appear in thinner silicon films. The variation of n_{eff} with thickness ratio is not surprising since the change of the thickness ratio alters the contribution of the silicon in the composite. Since we did not find a negative index in the simulation when the semiconductor layer is pure silicon, there must exist a critical value of impurity concentration N_c, above which a negative index in the composite can be observed. Therefore, it is important to select proper values of the impurity concentration and the thickness of silicon films when the ferrite material is determined.

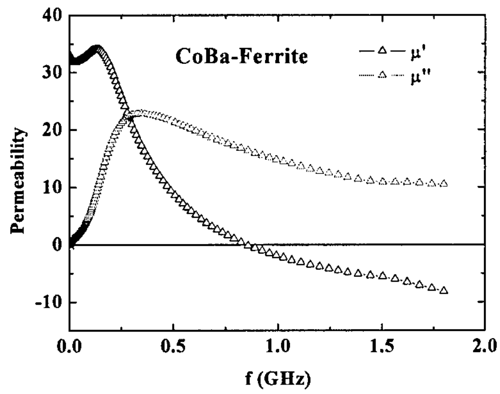

Figure 14, Microwave permeability of CoBa-Ferrite. Negative value was observed above 0.8GHz

Experimentally we have already demonstrated that negative permeability exists at CoBa-Ferrite (*Figure 14*). The permittivity can be designed with appropriate doping in semiconductor layer, which could be used to tune the negative index of refraction. Furthermore, it is also easier to fabricate compared to ferrite/metal multilayers since the thickness ratio between ferrite and semiconductor can be very reasonable.

3. **Design Criteria for DNM and SNM**

The ferrite/metal multilayers show simultaneous negative μ and ε, i.e. DNMs, leading to both left handed properties and negative index of refraction (NIM). On the other hand, ferrite/semiconductor multilayers show single negative μ, i.e., SNMs. Although NIM can be obtained, SNMs is not expected to possess other left handed properties. To unify these material systems and come up better

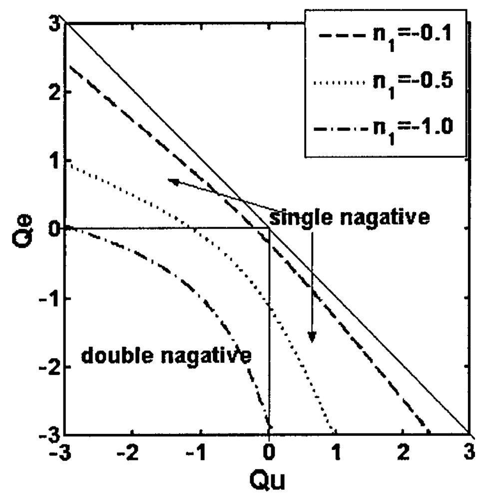

Figure 15, Index of refraction as a function of quality factors. Double negative materials (DNM) are NIMs and LHMs. However SNMs can be NIMs, provided appropriate values of quality factors.

design guide, we analyzed results and found all these properties can be represented as a function of quality factors $Q_\mu = \mu' / \mu''$ and $Q_\varepsilon = \varepsilon' / \varepsilon''$ as indicated in *Figure. 15,* which provide power designing guides for achieving NIMs.

4. Other material systems: Magnetic LSMO multilayers

Encouraged by these calculations and the fact that SNMs are much easier to achieve compared with DNMs, we naturally look for materials that of similar conductivity with semiconductors and more compatible with ferrite fabrication. Based on our prior knowledge on oxides showing colossal magnetoresistance (CMR) immediately become candidates. Resistivity of $(La_{1-x}Sr_x)_2MnO_4$ and other CMR materials fall into desirable resistivity values for NIMs discussed above. In addition their resistivity can be tuned with an external magnetic field, and most importantly, ferrites/CMR-material multilayer materials can be readily fabricated compared with E/M or ferrite/semiconductor multilayers. The resistivity tunability provides a convenient method to search for NIMs and eventually tune the properties of NIMs. The magnetic field will change the permeability spectrum since it changes FMR frequency.

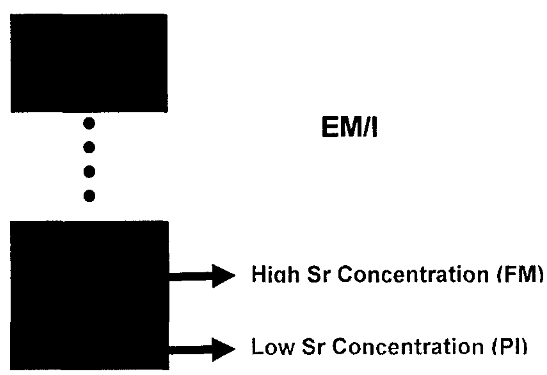

Figure 16, LSMO multilayer design
for m-LHM

Furthermore, CMR materials can be used to make NIMs themselves since they have very rich phases. We are particularly interested in two phase transitions, which are insulating-metallic transition and paramagnetic-ferromagnetic transition. Insulating-metallic transition gives field-induced negative permittivity and ferromagnetic phase gives negative permeability.

Figure 16 is the LSMO multilayer design for m-LHM. LSMO with high Sr concentration is ferromagnetic metal at room temperature, while it is paramagnetic insulator with low Sr concentration. Ferromagnetic metal gives both negative permeability and permittivity around FMR frequency. By inserting insulating layer between metallic layer, loss can be reduced dramatically. The advantage of this idea is those two layers have similar lattice structure, which makes sample ease to fabricate.

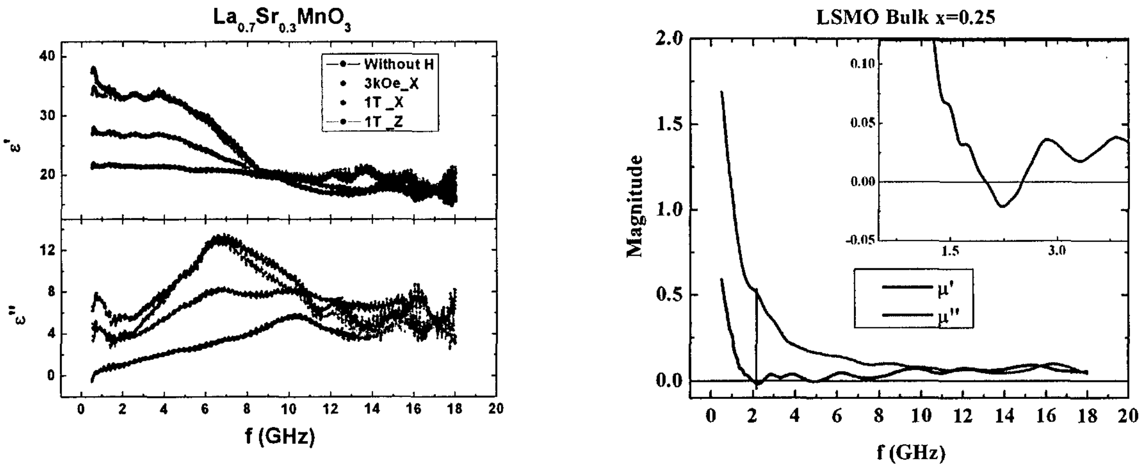

Figure 17, microwave permittivity and permeability of LSMO

Experimentally, we found the above design is very attractive. We observed negative permeability in LSMO bulk material and huge change of permittivity was realized by introducing magnetic field. The results are shown in *Figure 17*. It raises the possibility to achieve m-LHM by LSMO at room temperature.

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